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STUDY OF RADIOBARITES IN URANIUM MILL TAILINGS

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ABSTRACT

Scanning electron microscopy (SEM), energy-dispersive X-ray analysis (SEM-EDX), transmission-scanning electron microscopy (STEM), X-ray diffraction and X-ray fluorescence have been used in conjunction with a dispersed-grain, nuclear emulsion technique to search for radiobarites. In previous studies, other surface techniques including scanning electron microscopy, X-ray emission (SEM-XRE), secondary ion mass spectroscopy (SIMS), X-ray photoelectron spectroscopy (XPS, also called ESCA), were used but were unable to locate radiobarites.

In this present study, the dispersed-grain, nuclear emulsion technique has allowed us to locate radiobarites for the first time. The characteristics of the radiobarites were assessed in a preliminary manner, using SEM-EDX, TSEM and XRD, and selected area electron diffraction (SAED).

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1.0 INTRODUCTION

Chemical sequestering studies upon sulphuric acid leaching pachuca solids, and tailings from uranium mill tailings and thermodynamic analyses have suggested that radium-226 is primarily associated on the solid phase as a coprecipitate. (Constable and Snodgrass (1984), Snodgrass and Hileman (1985), Legeyt (1986). The most plausible solid phases controlling Ra-226 are barium sulphate and lead sulphate. The objective of this study was to build upon these past studies to attempt to identify the physical state of Ra-226.

2.0 CONCEPTUAL MODEL

Previous work (Stieff, 1985; IEC Beak, 1983; Constable et al, 1984; Wiles, 1978; Seeley, 1977; Benes et al, 1982; Nixon et al, 1982; Ryan and Levens, 1980; Yagnick et al, 1981; Levins et al, 1978; BEAK, 1985; Nirdosh, 1985; Nirdosh et al, 1984) suggests to us that the conceptual model given in Figure 2.1 presents a useful summary of the possible mechanisms by which Ra-226 may be coprecipitated in the leaching tanks. The model suggests

(i) that release of radium-226 from uranium tailings resulting from sulphuric acid leaching of the raw ore, is controlled by the dissolution of a parent crystal with which it is coprecipitated;

(ii) that Ra-226 is dissolved by acid attack on its parent uranium mineral, and reprecipitates near the uranium mineral or at a substantial diffusional distance away;

(iii) that this coprecipitate is formed in the first several leaching tanks (LP);

(iv) that a coprecipitation mechanism rather than reversible adsorption mechanism in the leaching tanks controls the partitioning of Ra-226 between the solid phase and the leaching liquor after dissolution of the parent U-238 mineral;

(v) that the parent crystal may be composed of sulphates of Ca, Ba, and/or Pb and that this crystal is formed on the surface of LP solids;

(vi) that this coprecipitate remains chemically stable in the tailings pile until the porewater concentrations of sulphate are low enough to allow dissolution of the mixed crystal containing Ra-226.

The sketch allows for a potential role for the fluid film thickness upon coprecipitation if fluid transport rather than surface reaction controls precipitation kinetics.

The conceptual model permits a framework for explaining and mathematically modelling the observation that, of expected levels of solution activity of the radioistopes, more

Ra-228 is found in the solution of controlled leaching experiments than Ra-226. A model of the Ra-226/Ra-228 differences would need to consider (i) whether the relative dissolution kinetics of Ra-226 from uranium containing minerals and of Ra-228 from thorium containing minerals are substantially different, (ii) the dissolution kinetics of barium and lead from their parent minerals, and (iii) their reprecipitation kinetics. Different dissolution kinetics for uranium and thorium minerals are a most plausible explanation of these differences. However, in a recent NUTP funded study (Legeyt, 1986), the rate of leaching of these two minerals (as measured by measurements of Ra-226 and Ra-228) in perchloric acid was similar (Steger, personal communication). Accordingly, only if sulphuric acid causes different leaching rates would dissolution kinetics explain the Ra-226/Ra-228 differences.

An overall kinetic model of all these processes is ultimately needed to prove the conceptual model outlined above. A possible set of equations to describe this conceptual model has been given in Snodgrass and Hileman (1985). The data required to substantiate and calibrate such a model are beyond the scope of this work. A few essential pieces of knowledge in evaluating the conceptual model are evaluated in this work. A few other essential steps are the subject of (i) other work with which members of this BEAK team are associated and funded as a strategic grant to McMaster University, and (ii) other projects funded by the National Uranium Tailings Program.

In the long-term, several steps will form the basis for substantiating or modifying, and mathematically describing the mechanisms postulated in Figure 2.1. They include:

(i) Identify the minerals from which radium, lead, barium and other elements of concern are derived, using standard mineralogical and micro mineralogical techniques.

(ii) Identify the solids formed, the species in solution, their mineralogical sources, and the concentrations of each over time as the ore dissolves;

(iii) Identify the fate of each of these solids and species in the process stream;

(iv) Identify the surface precipitate (host crystal) containing Ra-226 and Ra-228 with direct observational techniques and identify the cyrstallization site.

(v) Evaluate and quantify the kinetics of dissolution of pure minerals containing Ra-226, Ra-228, Ba, Pb, Ca, Fe and Sr.

(vi) Evaluate coprecipitation kinetics to explain Ra-226/Ra-228 differences in pure systems.

(vii) Evaluate steps (v) and (vi) in the heterogeneous mill system.

(viii) Couple steps (v), (vi) and (vii) with an evaluation of supersaturation conditions in the mill of the relevant parent crystal.

(ix) Validate that the same host solid controlling Ra-226 in the leaching pachua solids also controls its dissolution behaviour in old tailings.

(x) Trace the fate of each of the solids and soluble species in the mill processing streams during the history of the tailings behind their containment structure.

In this study, direct observational techniques (step iv) are used to identify a potential crystallization site for the host crystal of Ra-226. The preliminary nature of the study means that these techniques are used to demonstrate their ability to observe the host crystal. More work is required to identify, in a rigorous manner, crystal characteristics and to provide information required for experimental and modelling purposes.

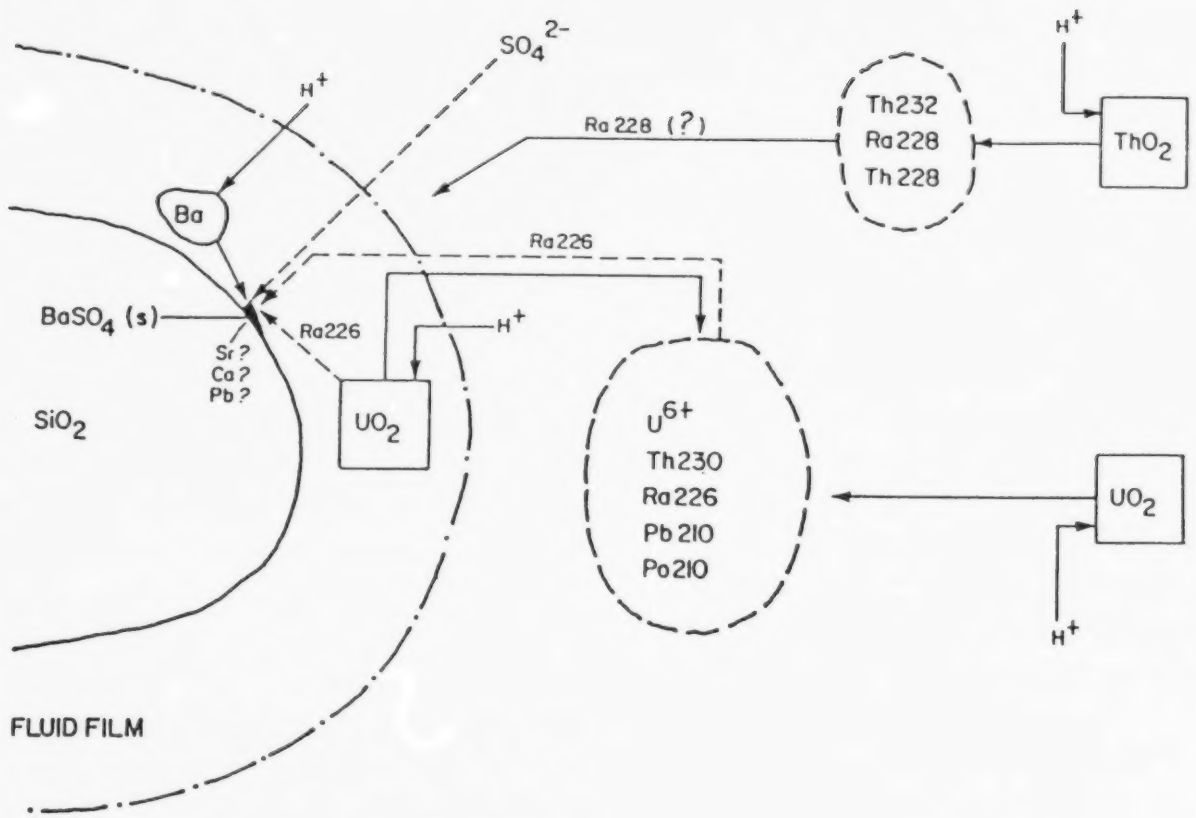

Figure 2.1 Conceptual Model (from BEAK, 1985)

3.0 STUDY METHODS

Previous studies have used surface analytical techniques (secondary-ion-mass spectrometry, SIMS; X-ray photoelectron spectroscopy, ESCA; Auger; scanning electron microscopy, SEM; Mossbauer) to examine the surface of the leaching-pachuca solids and of tailing samples. (Constable and Snodgrass 1984; Snodgrass and Hileman, 1985). The objective of their work was to identify the possible solids containing Ra-226. SEM studies showed small precipitates on the surface of the tailings of a thickness of 1-2 um. Because neither barium sulphate nor lead sulphate crystals were identified with SEM-XRE, it was hypothesized that these crystals must be less than 1 um in size. With Mossbauer, the presence of jarosite in tailings was identified, but not in the leaching-pachuca solids. The depth profiles with SIMS presented interesting data, but the beam averaged several particles because the beam width was 50-150 um. This is the typical beam width required for SIMS analysis. This width of beam precludes using the superior detection characteristics of SIMS for examining crystals of size 1-2 um. Further work is required in order to identify the location and morphology of $BaSO_4$(s) and $PbSO_4$(s) containing Ra-226.

In this study, it was hypothesized that a useful approach for identifying Ra-226 containing crystals was to use a technique which could identify Ra-226 alpha activity and then to examine the properties of the sites containing Ra-226 with other surface analytical techniques. At the concentration levels of Ra-226 on the solids, (5-10 Bq/g; or 10^{-10} mol percent), Ra-226 cannot be detected by such relatively sensitive surface techniques as SIMS. Autoradiography (e.g., Wiles, 1978) and dispersed-grain nuclear emulsion (Stieff, 1985) are two techniques capable of detecting the alpha emitters of U-238 decay series radionuclides. Autoradiography is non-specific in that it characterizes all sites containing alpha-emitting radionuclides (e.g., U-238, Th-230, Ra-226) whereas nuclear emulsion potentially has the ability to identify the specific radionuclide involved. The dispersed-grain nuclear emulsion technique was selected for this study.

The nuclear emulsion technique was first applied to U-mill tailings by Stieff (1985). It allows identification of Ra-226 associated with (i) radiobarites (tens of microns to fraction of micron in size), radio-colloids (colloidal-sized solids containing Ra-226), mineral coatings and adsorbed Ra-226. The technique is a micro-autoradiographic procedure that permits the direct observation of alpha activity of individual mineral grains or particles. The decay of Ra-226 and immediate (short-lived) daughters produce

a four branched star, a pattern which allows the identification of Ra-226. Ra-226 can be differentiated from other radionuclides (e.g., U-238, U-234, Th-232, Rn-222) because they produce different characteristic alpha tracks and/or stars. Coupled with the use of an optical microscope, SEM-XRE or other micro minerology techniques, one can identify the chemical precipitates with which the Ra-226 is associated.

These techniques have allowed Stieff to identify compounds such as (Ra, Ba) SO_4, (Ra, Ca) SO_4, (Pb, Pb-210) SO_4 and (Pb, Pb-210) CO_3. It is also possible that the technique can be used to identify the five-branch stars produced by Ra-228 and its daughters although this possibility was not pursued in this study.

The technique when fully evaluated, should allow us, to positively identify the physical state of Ra-226 on the surface of the solids "in an unequivocal fashion". It is a qualitative, and partially quantitative technique. When coupled with the chemical dissolution study, the appropriate distribution model for coprecipitation and the physical state of Ra-226 on the leaching pachucas solids can be well established.

The technique takes about 1-4 months for the alpha plate exposure. This allowed us to examine one set of solids in the time frame of this study. The ideal procedure is to set up two alpha plates per solids sample. One set is pulled early (after 1-1.5 months) to assess the density of alpha tracks. This inspection allows the experienced investigator to determine the ideal lapse time for examining the emulsions.

This study concentrated mainly upon using the nuclear emulsion technique. As a small component, some addition work with SEM was conducted using different sample preparation techniques.

Several tailings were selected for analyses by dispersed-grain nuclear emulsion including leaching-pachucas solids and uranium mill tailings. From these samples, two tailings materials were selected for analyses by dispersed-grain nuclear emulsion and characterization by surface analysis techniques. One was a barium sulphate sludge formed in the University of Alberta laboratory. The second was a sample of old Nordic tailings estimated to be approximately 30 years old. The old Nordic tailings were located on the surface of the pile for approximately a quarter of a century and then were continuously leached thereafter for five years in columns at the Wastewater Technology Centre, Burlington, Ontario. The specimen reported on herein was from the mid-depth of a high-rate leaching rate lysimeter (L8 of Constable and Snodgrass, 1984).

4.0 STUDY RESULTS

SEM Studies

As a part of this study, an aliquot of leaching pachucas solids from Rio Algom was examined by a SEM-EDX instrument with a beam capable of resolving 0.4 um size particles. The particles had been isolated by settling the leaching pachucas solids in an methylene iodide solution for density separation and then centrifuging the fine particles left in the supernatant after 1 hour settling. Density separation did not work well with the fine-grained material since surface tension effects become important. The objective of this approach was to obtain a sample of fines which might be enriched in small barium sulphate crystals. This examination is opposite from that of Atlantic Nuclear who examined the heavy fraction in a recently NUTP funded study (Steger, personnel communication).

A detailed search of different viewing fields showed that both whole fields and specific particles were dominated by aluminum and silica. Some specific particles were enriched in iron and sulphur.

A search was also made of the particles using the backscatter scanning mode. In this mode, areas on the surface composed of high molecular weight elements are identified as white spots on a dark background. After some searching, several concentrated areas of white spots were identified. Analysis of these spots under EDX initially suggested a barium peak with no sulphur peak. Further inspection of the SEM-EDX software showed that the peak was actually a titanium peak which coincides very closely with the primary barium peak. Hence with such samples, identification of a barium peak must be checked against titanium peak. Continued searching with the SEM did not reveal any areas which could be identified as containing barium.

Nuclear Emulsion Results

A barium sulphate sludge was examined with the intention of using the track density in the nuclear emulsion and the known Ra-226 content of the sludge to calibrate the nuclear emulsion method for application to tailings material. The barium sulphate "sludge" produced only a few four-track stars which are characteristic of Ra-226, despite the

analyses received from the University of Alberta which suggested that the Ra-226 content was about 100-1000 Bq/g. The density of stars was judged by the analyst to be too low for the claimed value of Ra-226 activity. The nuclear emulsion showed the presence of many single tracks which are characteristic of Pb-210 and Po-210.

The sample was analyzed by XRD and confirmed as primarily barium sulphate. It also contained a fine-grained aluminum-bearing material (by SEM analyses) which the analyst suggested was an aluminum hydroxide or other hydrated aluminum oxide that did not appear in the XRD analyses. No other chemical elements such as uranium or thorium decay products were observed.

The barium sulphate "sludge" was inserted into a gamma counter and the spectrum area of the 609 keV peak for Bi-214 counted in a cursory manner. The sample had been inserted into the vial and sufficient time allowed for Bi-214 ingrowing from Ra-226 (and Rn-222) to occur. The count for Bi-214 suggested that some Ra-226 was present in the sample but at a much lower level than expected. Further work is required to resolve this conflicting evidence. It may simply result from mislabeling of a sample. The samples involved have been submitted for Ra-226 analysis. This problem points out the need to develop standards for counting and evaluating alpha stars.

The tailings material produced alpha-stars which are characteristics of Ra-226. These particles were examined under optical microscope and then representative particles further analyzed using SEM-EDX, STEM and SAED techniques.

Three track-producing particles of size 10-30 microns were picked from the emulsion for surface analysis. Figures 4.1-4.3 are optical photomicrographs of the emulsion, after the particles were removed. Figures 4.4-4.6 are SEM photomicrographs of the three particles. These are back-scattered electron photomicrographs and, consequently, the higher atomic number material appears brighter in the image. Figures 4.7-4.9 are x-ray spectra of the three particles. Particles #1 and #2 are silicates, but particle #3 is barium sulfate. The silicates (Figures 4.7; 4.8) are composed primarily of silicon but contain some aluminum. Figure 4.8 (Particle 2) indicates that some barium is also present - the barium peaks are judged to be significant (compared to background) and are not titanium. The bright spots shown on Figure 4.4 are the source of the barium in the spectra for Particle 2 (Figure 4.8).

Further analysis of the bright areas on Particles 1 and 2 was made. Figures 4.10 and 4.11 are x-ray spectra of the small, bright crystallites shown in Figures 4.4 and 4.5. The x-ray spectra were made by focusing the beam upon one small crysallite (bright spot) on the parent particle. The beam, in this mode, encompassed an area about 1-2 um in width. This gives fairly precise information on the composition of the crystallite. Figures 4.12 and 4.13 are higher magnification SEM photomicrographs of these crystallites shown respectively on Figure 4.4 and 4.5. The x-ray spectra indicate that the crystallites contain barium, sulfur and lead. The higher magnification indicate that there is a large quantity of submicron sized particles which appear as "bright spots" and which contain the barium, sulfur and lead. Figures 4.12 and 4.13 also provide some qualitative insights into the type of crystal sites upon which these submicron crystals form and grow. These insights need to be developed further before they are applied to the modelling steps described above.

The submicron crystallites on Particle 1 were removed from particle 1 by crushing particle 1 and then dispersed onto a thin carbon film for scanning transmission electron microscope (STEM) analysis. Crushing caused the small crystallites to be freed from the parent silicate and thus allowed the analyst to examine the crystallites by STEM independent of the parent silicate. Figures 4.14 and 4.15 are STEM photomicrographs of the dispersion. In these photos, the higher atomic number containing material appears darker.

Selected area electron diffraction (SAED) analysis of the barium, lead and sulfur-containing crystallites was also made. SAED results indicated that the particles are barium sulfate probably in solid solution with lead sulfate. We cannot unequivocally conclude that the lead is in solid solution with the barium sulphate, but the SAED analysis indicates that there was only one phase (a necessary condition for solid solution) not two separate phases.

The submicron crystallites are primarily barium sulphate. A partially quantitative estimate of the Pb composition was made with the SAED analysis but requires further work to make it adequate for communicating in report form.

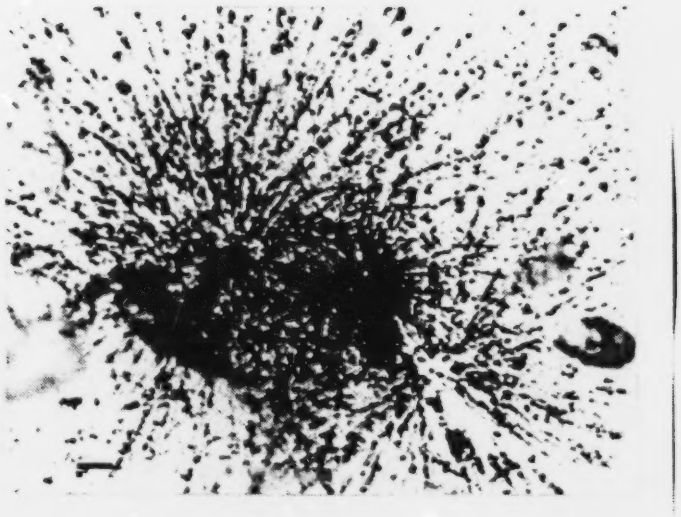

Figure 4.1 Optical Photomicrograph of Alpha Tracks in Nuclear Emulsion After Removal of Particle 1. Sample is from old Nordic Tailings (Magnification: 750 times. Bar = 10 um)

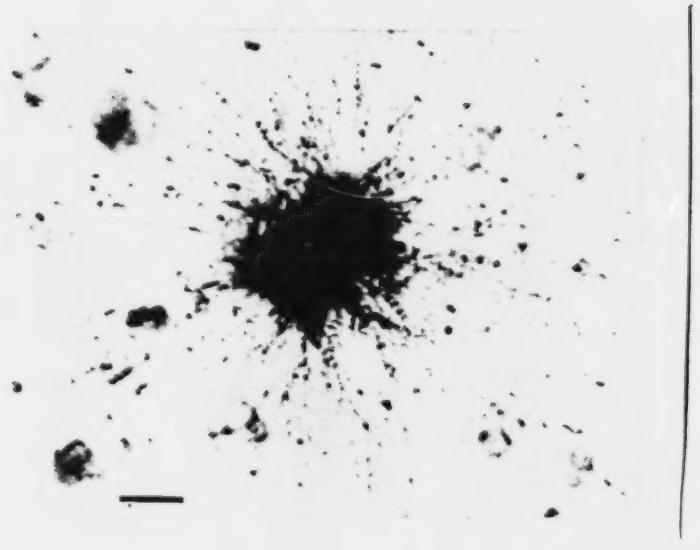

Figure 4.2 Optical Photomicrograph of Alpha Tracks in Nuclear Emulsion After Removal of Particle 2. Sample is from old Nordic tailings (Magnification: 750 times. Bar = 10 um)

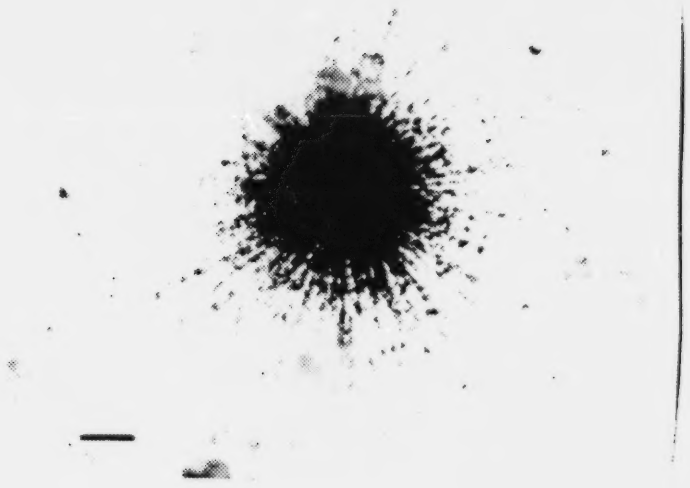

Figure 4.3 Optical Photomicrograph of Alpha Tracks in Nuclear Emulsion After Removal of Particle 3. (i.e., Large barium sulphate crystal) from old Nordic Tailings
(Magnification: 750 times. Bar = 10 um)

Figure 4.4 SEM Photomicrograph of Particle 1
(Magnification: 600 times)

Figure 4.5 SEM Photomicrograph of Particle 2
(Magnification: 1200 times)

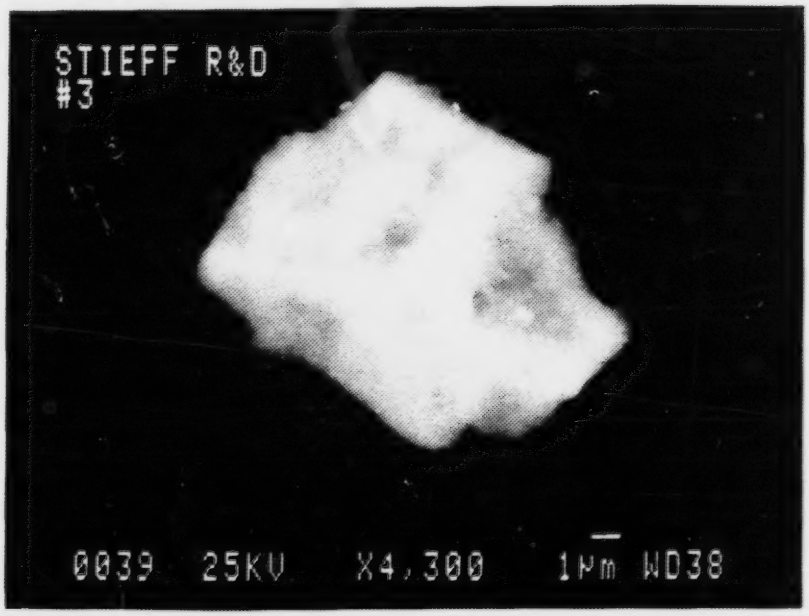

Figure 4.6 SEM Photomicrograph of Particle 3. Large barium sulphate crystal
(Magnification: 4300 times)

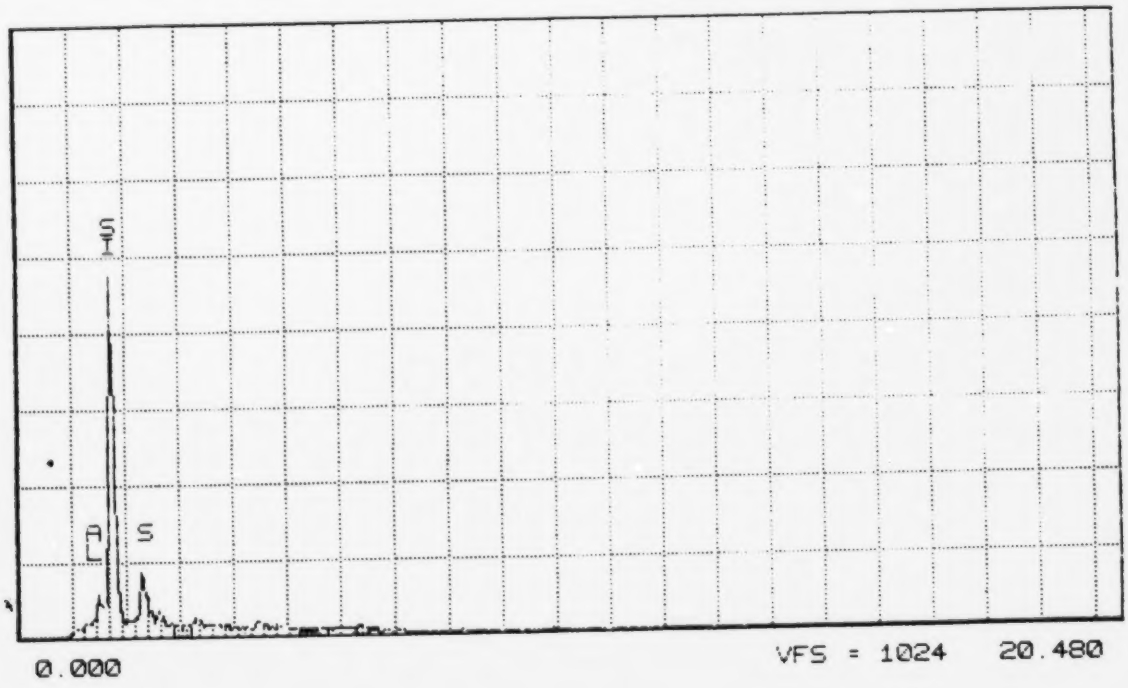

Figure 4.7 SEM-EDX Spectrum of Particle 1

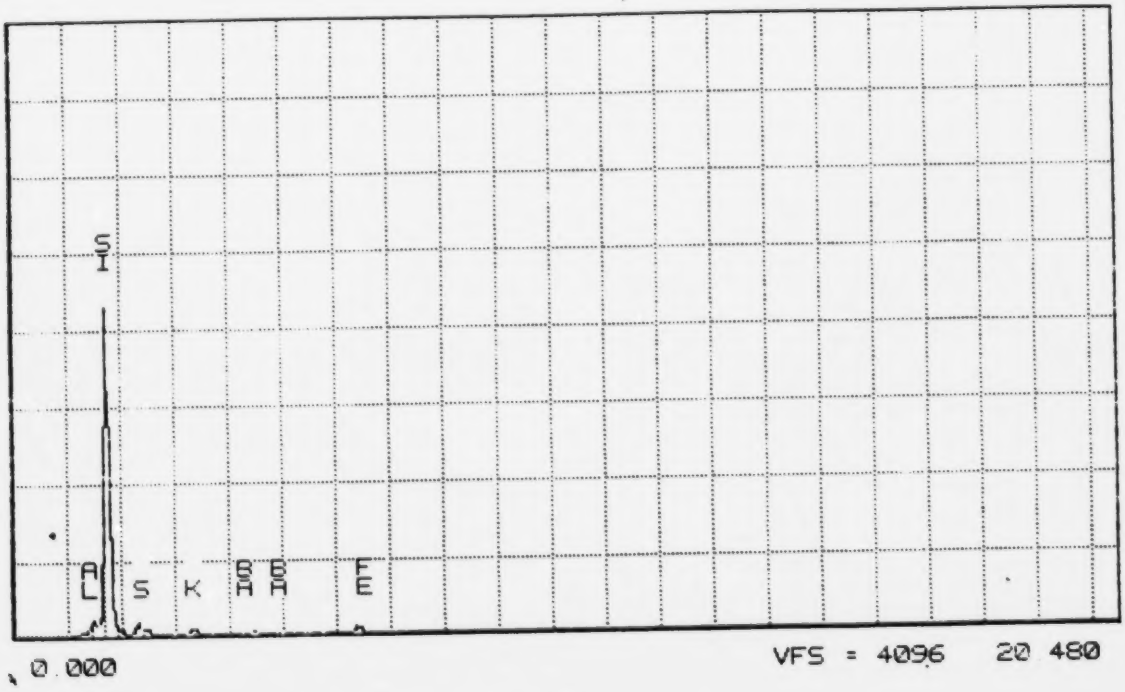

Figure 4.8 SEM-EDX Spectrum of Particle 2

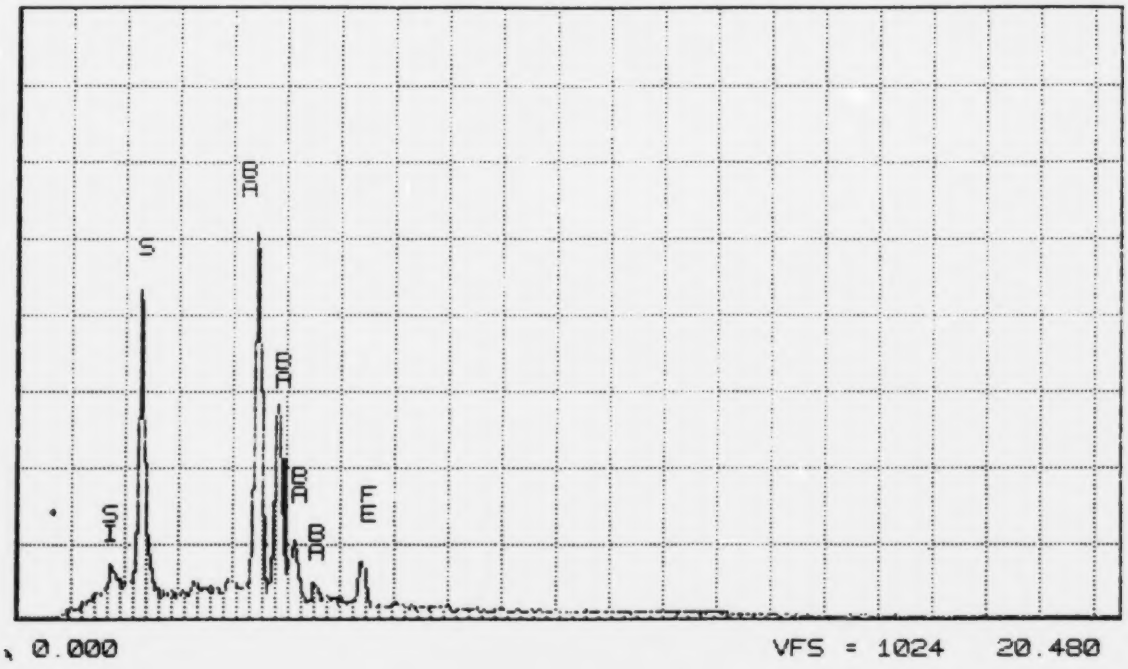

Figure 4.9 SEM-EDX Spectrum of Particle 3

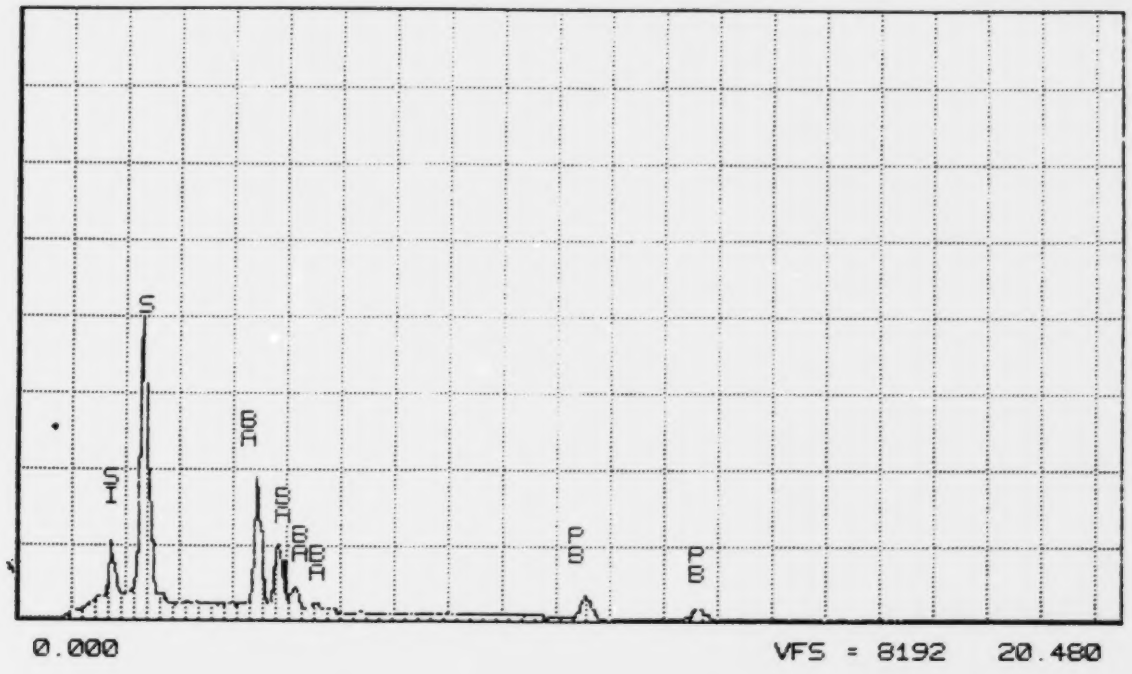

Figure 4.10 SEM-EDX Spectrum of Crystallite on Particle 1

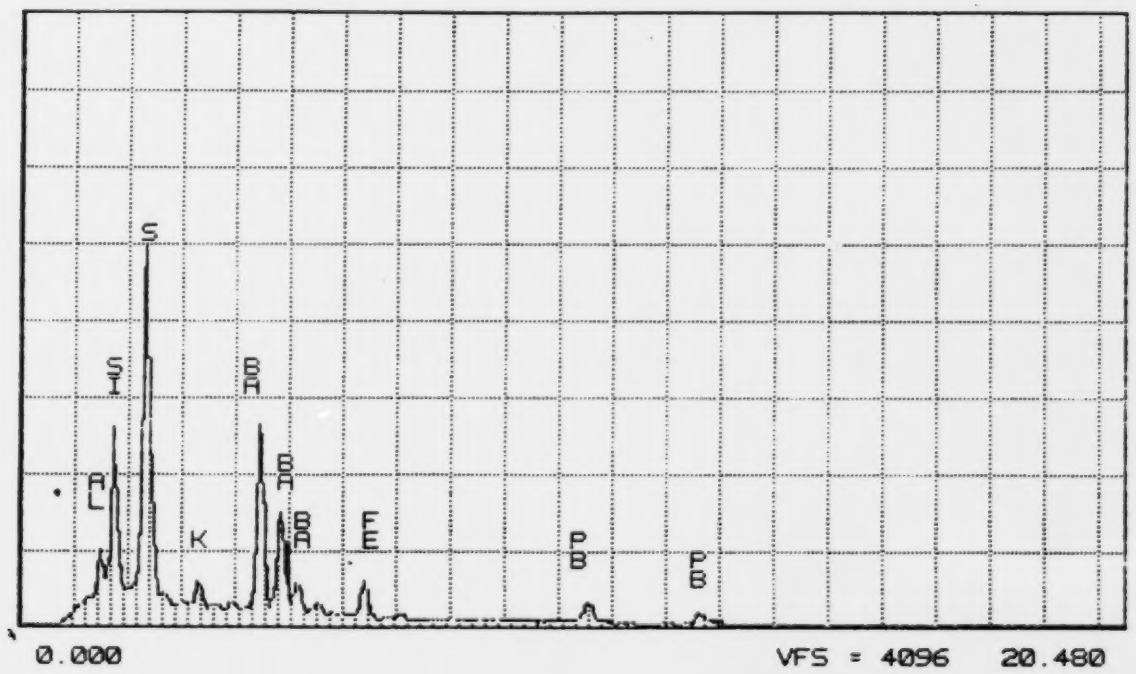

Figure 4.11 SEM-EDX Spectrum of Crystallite on Particle 2

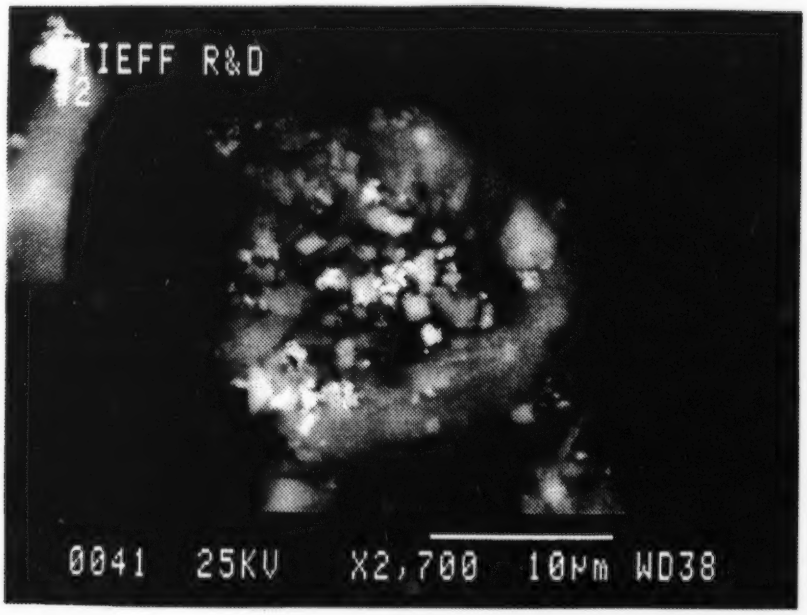

Figure 4.12 SEM Photomicrograph of Crystallites on Particle 1
(Magnification: 2700 times)

Figure 4.13 SEM Photomicrograph of Crystallites on Particle 2
(Magnification: 800 times)

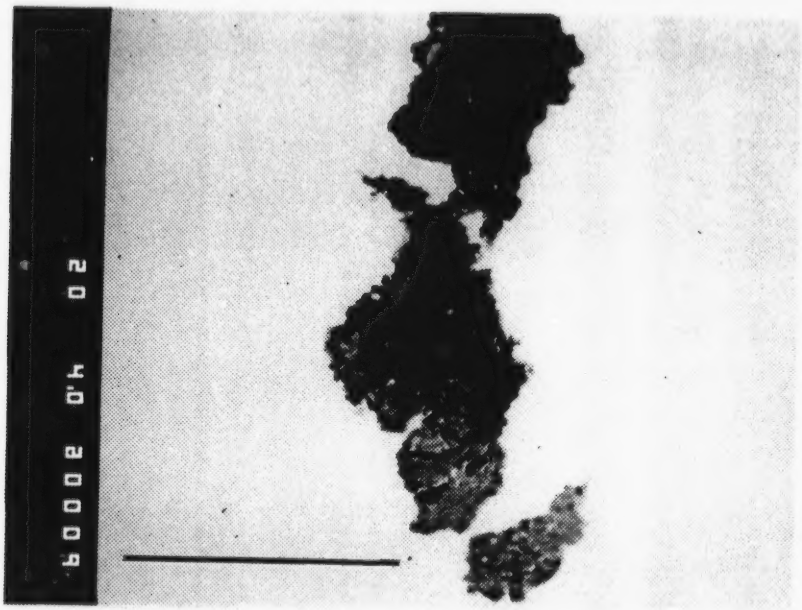

Figure 4.14 STEM Photomicrograph of Crystallite Dispersion of Small Particles Removed from Particle Number 1
(Magnification: 4000 times. Bar = 10 um)

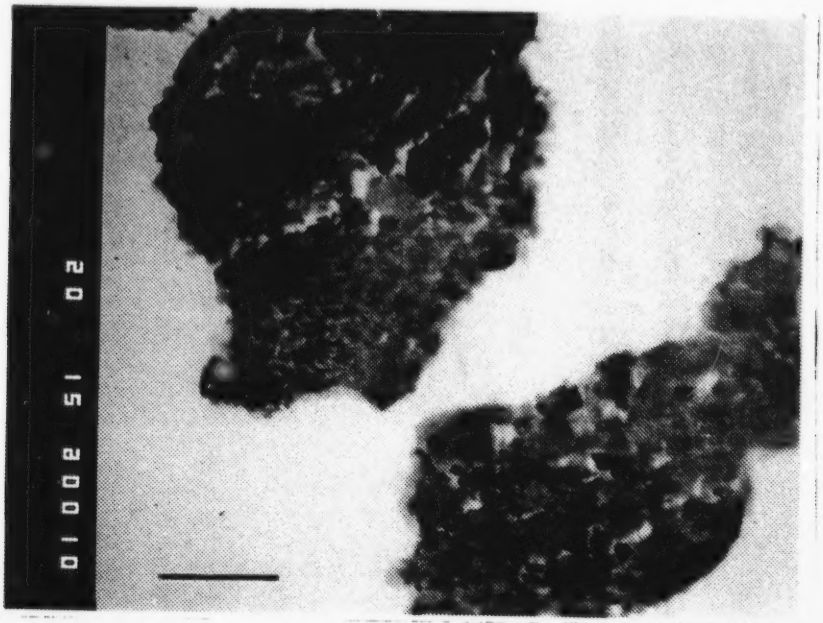

Figure 4.15 STEM Photomicrograph of Crystallite Dispersion of Small Particles Removed from Particle Number 1
(Magnification: 1500 times. Bar = 1 um)

5.0 DISCUSSION

This study had the primary objective of evaluating the ability of dispersed-grain nuclear emulsion and surface techniques to identify barium sulphate and lead sulphate crystals containing Ra-226. These crystals are of primary interest because past dissolution studies and thermodynamic analysis have suggested that Ra-226 is primarily present in tailings in these crystals.

This study demonstrates that the nuclear emulsion technique can be used to locate barium sulphate particles containing Ra-226 and lead. Application of the nuclear emulsion technique aids SEM technology in identifying barium sulphate particles, because the barium content of the tailings on a bulk basis (per gram for example) is at the SEM detection limit.

This work has isolated two types of barium sulphate particles. One large particle of the order of 10 um in size was found and is essentially composed of barium sulphate. Several submicron size particles have been found and analyzed using SEM-EDX, STEM and SAED; they are composed primarily of barium sulphate but also contain lead which appears to be in solid solution. Further examination of the micrographs would give the reader some insights into the type of site upon which they are located. Provided it can be shown that these small crystallites formed there, this gives insights into the type of mineral upon which crystal growth occurs.

One interpretation of the two types of particles is as follows. The submicron particles were formed initially in the mill. The small particles would typically be formed with relatively quick kinetics in the leaching pachucas. The lead content would make the small particles thermodynamically unstable relative to pure barium sulphate. The effective solubility of barium with respect to these particles is also higher than 10^{-10} M^2, the solubility product of pure barium sulphate (at ambient environmental temperature). Hence the small particles would dissolve first but barium could crystallize in a purer barium sulphate form. The possible site of barium crystallization is the large 10 um barium sulphate particle. For a well formed shape, and its large size, slow kinetics are expected to dominate its formation. Such kinetics are possible for the time scales of the tailings site.

In summary, the interpretation is that the small barium sulphate particles containing lead and Ra-226 are formed in the sulphuric acid leaching tanks. But they subsequently dissolve due to their thermodynamic instability. Then barium reprecipitates as a purer barium sulphate crystal and coprecipitates radium-226.

There is one problem with the interpretation. The barium photomicrograph shows a rounded pit suggesting that the crystal was dissolving. There are two possible explanations. (i) The crystal was epigenetic being derived from the ore, or was formed in the leaching tanks. (ii) The crystal was formed in the tailings pile but then subsequently started to dissolve. The latter explanation (tailings pile formation) may be possible because the sample is derived from a lysimeter in which Ra-226 was bleeding from the lysimeter at a low activity for approximately 5 years but after which a large peak Ra-226 activity occurred just before the lysimeter was shut down.

But it is most probable that the large barium sulphate crystal is epigenetic. In another NUTP study, Atlantic Nuclear Services attempted to isolate the radiobarites by liquid separation, but collected the heavy fraction (Steger, personal communication). They used SIMS to detect the presence of primary barium sulphate containing from 100 ppb to 5 ppm radium-226 in particles from 150 to 20 um in size. To date they have looked at over 40 such particles. Particle #3 is likely one such particle, i.e., epigenetic.

The epigenetic barium sulphate crystals and barium contained in sericite (Jonasson, personal communication) would represent sources of barium in the ore for the coprecipitation process of interest. It would be useful in future studies to determine dissolution kinetics of these and other barium containing minerals to ascertain the main source of barium for co-precipitation.

In the future, it would be useful to pursue several lines of inquiry. These are detailed in the next section. Two others are amplified herein: other samples, and the role of lead.

It would be useful to apply the techniques of this study (i) to leaching pachuca solids before addition of neutralization chemicals but after leaching, and (ii) to solids which have been experimentally washed with various sequestering agents (e.g., in Snodgrass and Hileman, 1985). This would aid definition of the attributes of the crystallites before they are substantially weathered. The solid sample, studied in this project (Nordic and then

Environment Canada, Burlington lysimeter), has been weathered. There is some question about relating the data to geochemical events. However, the nature of the treatment in the CCIW lysimeters does not overly complicate the interpretation because the experimental manipulation involved addition of distilled water at a rate higher than normal rainfall. No other experimental manipulation was used. Hence, the time history of the sample is essentially one of pyrite oxidation and aqueous dissolution due to infiltrating water in the saturated zone.

With respect to lead, no particles containing radium-226 and composed primarily of lead sulphate were observed in this study. The observed particles had lead contained within a barium sulphate matrix. But a recent study (Legeyt, 1986) suggests that under conditions of repeated washing with distilled water, most of Ra-226 dissolution is initially associated with barium sulphate dissolution during the first few days and thereafter with lead sulphate dissolution. This suggests that future work should include a search for separate lead sulphate crystallites containing Ra-226.

6.0 FURTHER WORK

This work has demonstrated that the nuclear emulsion technique can be used to identify the site of Ra-226 containing crystals. More work is required to identify the physical state of Ra-226 in an unambiguous way. Such work includes

(i) Do a more detailed analysis of the old Nordic tailings to ascertain the relative population balance of submicron barium sulphate (containing lead) crystals and the large 10 um sized barium sulphate crystals.

(ii) Carry out further studies to identify the relative number of Ra-226 containing crystals and Pb-210 (Po-210) containing crystals in the old Nordic tailings samples.

(iii) Carry out studies upon ore samples and leaching pachucas solids used in previous NUTP funded work to identify the characteristics of crystals containing Ra-226 and to search for epigenetic barium sulphate crystals.

(iv) Examine the substrate containing the submicron barium sulphate crystals to identify the site of crystal growth. (This information is required to evaluate crystal growth theory for applicationto the leaching tanks).

(v) Quantify the composition of other elements (e.g., Pb) in the barium sulphate crystals.

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